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Mossbauer study of cation substitution effect on the magnetic phase transition temperature of $0.5AFeO_3$-$0.5NaNbO_3$ (A = Bi, La) solid solutions

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$BiFeO_3$ is the most widely studied multiferroic due to high temperatures of both ferroelectric and magnetic phase transitions. Recently a possibility of magnetic superexchange between Fe^{3+} ions in $BiFeO_3$ via the empty 6p states of Bi^{3+} was predicted theoretically [1]. Earlier we showed the role of Pb^{2+} ions which also possess the empty 6p states, in the enhancement of the T_N in perovskite $Pb_{1-x}A_xFe_{1/2}Nb_{1/2}O_3$ (A = Ca, Ba) solid solutions [2, 3]. However this mechanism of superexchange seems to become noticeable only at high enough dilution of the Fe-sublattice, i.e. in solid solutions.

The $0.5AFeO_3$-$0.5NaNbO_3$ (A = Bi, La) solid solution compositions were prepared by solid phase reactions route from high-purity oxides. The XRD study revealed that the samples are single phase and perovskite structure. Room temperature Mossbauer spectra of both show the quadrupole-split lines. The main reason of quadrupole splitting in such systems is compositional disorder in *B*-sublattice. All spectra contain 2 or 3 doublets with different values of quadrupole splitting. The isomer shift values of doublets correspond to the Fe^{3+} ions in oxygen octahedron. The presence of 2 or 3 doublets indicates that Fe^{3+} has 2 or 3 different environments in the lattice which may be a fingerprint of *B*-cations short-range ordering or clusterization [4].

To estimate the temperature of magnetic phase transition (T_N) we performed the measurement of Mossbauer spectrum line intensity under subsequent temperature lowering. Near magnetic phase transition the Mossbauer spectrum transforms from doublet into sextet. This transformation is accompanied by a dramatic decrease of Mossbauer spectrum line intensity I_m. Thus, the abrupt drop in the $I_m(T)$ dependence corresponds to the T_N. Mossbauer studies have shown that while for $0.5BiFeO_3$-$0.5NaNbO_3$ the T_N value is about 150 K, for $0.5LaFeO_3$-$0.5NaNbO_3$ it is only ≈ 20 K. This dramatic difference in T_N values seems to be due to additional contribution of the superexchange between Fe^{3+} ions via the empty 6p states of Bi^{3+} to the overall superexchange in accord with predictions of De Sousa et al. [1].

For both compositions magnetization *M* studies in the 5-200 K range revealed only a maximum at 20-25 K in the zero-field cooling mode. In the field-cooled mode this maximum was not observed. Such behavior is typical of spin glasses. In $PbFe^{3+}_{1/2}B^{5+}_{1/2}O_3$ (B^{5+} = Nb, Ta) perovskite multiferroics a spin-glass state is known to coexist at low temperatures with the antiferromagnetic state [2, 4]. The absence of the *M(T)* anomaly at around 150 K for $0.5BiFeO_3$-$0.5NaNbO_3$ composition seems to be due to the strong diffusion of this anomaly. Similar "disappearance" of the M(T) anomaly corresponding to antiferromagnetic phase transition was observed, e.g. for some compositions of the $PbFe_{2/3}W_{1/3}O_3$-$PbTiO_3$ and $PbFe_{1/2}Nb_{1/2}O_3$-$PbTiO_3$ solid solutions [5, 6].

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